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AEROSOL STUDIES AT UMIST

Final Technical Report

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Department of Pure and Applied Physics
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July 1980

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M. J. Gay
M J Gay
Research Scientist

J. Latham
J Latham
Principal Investigator

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The first part of this report details a review of the dispersion of pollutants by rainfall, with particular reference to inertial and diffusive removal by precipitation, inertial, condensational and diffusive removal by cloud drops, and the influence of electrical forces on scavenging. The second half of the report describes a preliminary study of the influence of meteorological parameters on the size distribution of natural aerosol particles, with particular reference to two data		

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sets obtained in urban and rural localities. The size spectrum investigated lay between 0.5 and 10 ^μ equivalent particle diameter and the correlation between particle concentration and wind speed, relative humidity, temperature, atmospheric stability and rainfall were determined for various size categories.

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1. INTRODUCTION

Much of the work described in this report constitutes an extension of the preliminary studies carried out in the three years prior to December 1976 and described in the Final Technical Report under contract no. DA-ERO-124-74-G0023. Two main laboratory studies undertaken during the period April 1977 - April 1980, to examine the scavenging of submicron particles by electrified drops and the influence of mixing on the evolution of droplet spectra, have been reported in Interim reports and we include in this report some preliminary studies of the dispersion of pollutants by rainfall.

From studies made in the UMIST laboratories and at the field station of the Atmospheric Physics Research Group at Great Dun Fell, Cumbria, we describe the results of an investigation of the influence of meteorological parameters on the size distribution of natural aerosol particles in both urban and rural environments.

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2. DISPERSION OF POLLUTANTS BY RAINFALL

The Earth is 40% covered by clouds: this figure being arrived at by average albedo measurements. The efficiency of cloud removal processes is high over a wide range of aerosol particle sizes in contrast to the processes occurring below cloud which mainly occur for giant particles (larger than 2 microns). Wet removal is usually divided into rainout (in-cloud processes) and washout (below-cloud processes). Evaporation below cloud base will increase the concentrations of the constituents and the presence of high wind will affect the number of drops removed.

2.1 Inertial removal by precipitation

Below cloudbase sweepout of giant aerosol particles in the range (2 to 10) microns is the major removal process. Although this process may occur in clouds, the majority of the time the droplet is in flight it is below cloud base. At the 500 millibar level, raindrops travel at 50% higher velocities than at ground level (Beard 1976).

Sweepout involved the collection of particles in a raindrop's path. This collection depends on the inertia of the particle and the Reynolds number of the flow around the drop. The fraction of particles captured by the drop to the particles beneath the drop is the collection efficiency E .

Assuming unity collection efficiency, the area swept out depends on the area of the drop (r^2), whilst the volume of the drop (r^3) determines the net concentration. Thus collection of particles below cloud base would result in $K \propto \frac{1}{r}$ where K is concentration of pollutant in drop and r is radius of drop. Evaporation increases the relative concentrations in the smaller drops. Turner (1955) found for freezing clouds that $K = r^{-x}$ where r is radius of drop and K the concentration. x was found to be variable and he attributed this result to evaporation and

collection of salt below cloudbase.

According to Junge (1963) the net concentration in the drops (K'') is given by

$$K'' = fK + K'$$

$$K' = \frac{nH}{r} E a^3 N(a) da$$

f is a term to represent evaporation

K' is the washout term

a is the radius of an aerosol particle

r is the radius of a typical raindrop

E is the collection efficiency

H is the height of fall below cloudbase

K represents rainout

N is the number density of particles radius a .

The collection efficiency E has been calculated empirically by many workers from Langmuir (1945) onward, e.g. Beard and Grover (1974). These calculations were based on inertial deposition on a drop assuming unity coalescence efficiency. However, this is only the case for hygroscopic particles and some of these will already have been removed preferentially incloud and by other drops.

Ranz and Wong (1952) measured the collection efficiencies for spheres as a function of Stokes' number. They found the simple relation

$$K = Stk = \left\{ \frac{2pa^2}{9n} \right\} * \left(\frac{V}{r} \right)$$

E is collection efficiency

Stk is Stokes' number

p is density of aerosol particle

V is terminal velocity of raindrop

n is viscosity of air

Other parameters as previously defined.

The function $\frac{V}{r}$ does not change very much with radius. However,

the a^{-1} dependence of the collection efficiency means that the variation of this with aerosol particle radius will be a step function. Thus particles smaller than 1.0 micron will hardly be removed but particles larger than this will all be removed. Storebo and Dingle (1974) looked at pollution removal theoretically and came to the conclusion that hygroscopic particles were easily removed. Increased windspeed was shown to give a decade's difference in concentrations at 25 kilometres, probably because of increased condensation rate and less removal of drops.

2.2 Removal by diffusion to precipitation

Diffusive removal by precipitation is treated by the advection-diffusion equation

$$\frac{dn}{dr} = D^2 n - v n$$

n is number of particles per unit volume

v is velocity of fluid

D is diffusion constant

(See Twomey 1977). The collection efficiency is specified by the Reynolds number $\frac{\rho r V}{\eta}$ and Peclet number $\frac{2rV}{D}$

ρ is density of fluid

r is radius of raindrop

V is terminal velocity of drop

D is diffusion constant

Friedlander derived the expression (1967)

$$E = 4 Pe^{-1} + (1 + 0.2 Re^{1/6} Pe^{1/3})$$

E is diffusion collection efficiency

Pe is Peclet number

Re is Reynolds number

The expression $(0.4 Re^{1/6} Pe^{1/3})$ varies weakly with Reynolds number and is negligibly small, hence it can be deduced that

$$E = \frac{20}{rV}$$

so that for an order of magnitude change in drop radius (e.g. from drizzle to raindrops) the efficiency is changed by approximately a factor of 10. Although the efficiencies of removal appear very low, the numbers of particles in the lower size ranges means that considerable amounts of aerosol will be removed, e.g. below 0.1 microns. For raindrops and particles 0.4 microns the efficiency is 10^{-5} and this is very close to the experimental limits of measurement.

The intermediate range between diffusion and inertial removal (0.1 to 1.0 microns) has been investigated by Kerker and Hampl (1974) who found experimentally that Zebel's expression was valid:

$$\epsilon = 1.68 Pe^{-2/3} \text{ which is in the range } 10^{-4} \text{ to } 10^{-5}.$$

2.3 Inertial removal in cloud

The terminal velocity of cloud drops is small (1cm per second). Falling cloud drops are not very effective at scavenging smaller particles even though coalescence into raindrops involves a very large number (about 1 million) of cloud drops.

In the case of clouds which comprise ice crystals, e.g. cumuliform types, the ice crystals may act as scavengers of aerosol particles. Pitter (1977) has numerically modelled the scavenging of aerosol particles by thin plates under atmospheric conditions of -18°C , 400 millibars, ice crystal lengths being 103 - 366 microns. The model included hydrodynamic, gravitational and electrostatic forces. He concluded that for 103 microns crystals, scavenging efficiencies of 0.05 to 0.4 for aerosol particles from 0.5 to 10 microns. He found that for uncharged ice crystals, there was a cut off at 5 microns. This also applied to repulsive charges. Sood and Jackson (1970) investigated

experimentally snow scavenging for particle sizes 0.25 microns, 0.5 microns and came to the conclusion that there is a pronounced minimum in scavenging efficiency at about 0.4 microns. Scavenging efficiencies of the order 0.6 to 2% were found for sodium chloride aerosol particle sizes 0.1 to 0.16 microns.

2.4 Removal by nucleation

Cloud drops will begin to grow when a critical supersaturation has been maintained. A typical hygroscopic particle acting as condensation nucleus will undergo considerable dilution as the drop grows from critical radius (0.02 microns) to a typical cloud drop size of 10 microns. Katz and Kocmond (1973) concluded that cloud condensation nuclei may be considerably larger. According to Rogers (1976), typical droplet number densities lie between 50 and 200 per cc although aerosol size distributions are very variable only a small number of particles serve as cloud condensation nuclei, e.g. for the Continental size distribution of Junge (1963) 10% serve as nuclei. Following nucleation, the resulting cloud drop has sufficient inertia to grow by condensation and accretion to be removed gravitationally. If a cloud formed by condensation of the largest drops on the largest cloud nuclei the subsequent growth by coalescence to raindrop size would occur relatively quickly. Turner (1955) applied this argument to non-freezing clouds in Hawaii to account for the minima found in his results. Coalescence is still an important growth mechanism for cloud drops in warm clouds with freezing tops (see Rogers, 1976). Takahashi (1976) developed a computer model for warm clouds which correlated with Turner's results for Orographic Hawaiian rain.

2.5 Diffusion to cloud drops

Brownian diffusion of primarily Aitken particles to cloud

drops is a very efficient removal process in the size range

less than 0.05 microns. Junge (1963) has shown from the Smoluchowsky formula that:

$$\frac{dn}{dt} = 4n d r_c n_c \quad \text{gives a half life of}$$

$$T = 0.69/4n d r_c n_c \quad \text{where}$$

n = concentration of Aitken particles

d = diffusion constant of Aitken particles

r_c = radius of cloud drops

n_c = number of cloud drops

t = time

and he goes on to show for $r_c = 10$ microns and $n_c = 200$ per cc (Weickmann and Kampe 1953) corresponding to a liquid water content of 0.84gm per cubic metre

r microns	0.02	0.03	0.1
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T hours	0.64	3.8	38
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The estimate will not change much if charges are considered.

This treatment is justified since the separation of drops is about 1mm so that there is no interaction of the drops. The raindrops formed by coalescence of these cloud droplets will all have the same concentrations of contaminants. Goldsmith, Delafield and Cox (1963) have shown that this process is very efficient for particles smaller than 0.02 microns.

$$w_{BD} = \frac{3D p_t p_a}{a^2 p_w} \quad \text{where}$$

w_{BD} is the washout coefficient defined by

$$w_{BD} = \frac{\text{specific activity of rain}}{\text{specific activity of air}}$$

in the case of the radioactive scavenging studies. Electric charges were assumed to have negligible effect in the presence of ionising radiation.

D is the diffusion constant

p is the density of aerosol

p_a is the density of air

ρ_w is the density of water

a is the radius of aerosol particles

t is time

The time scale is quite significant e.g. for a duration of 10 hours all the particles radius 0.02 microns and most of 0.03 microns will be removed. The flux of aerosol particles along the vapour gradient (Facy effect) may be important for particles smaller than 0.1 microns. Goldsmith (1961) obtained low rates of attachment, though this took no account of electric charges.

Phoretic forces, produced by ordered fluxes superimposed on the relatively disordered motion in a gas are thought to contribute very little to scavenging. Diffusiophoresis is caused by water vapour produced in connection with evaporation and condensation. Thermophoresis occurs similarly. However, the integrated flux is negligibly small for the condensation processes only last for 10 seconds of a typical cloud drop's life. For particles in the sub-micron range, the effect was shown to be negligibly efficient by Goldsmith, Delafield and Cox (1963), typically 0.1 - 0.5%. Slinn and Hales (1970) have investigated the role of phoretic forces in the scavenging of particulates in the size range 0.01 to 1.0 microns and found efficiencies in the range 0.0005 to 0.01 for thermophoresis for raindrops 0.1mm to 1mm. Diffusiophoresis was about one third as efficient. These processes may occur while droplets are growing before coalescence becomes the main removal process. Using radioactive tracers, Perkins, Thomas, Young and Scott (1970) found that light rain takes longer to develop than heavy rain. They also found that rainout was initially high, then decreased during a thunderstorm and towards the end of the thunderstorm would increase again. They concluded that crops may be subject to several cycles and that in heavy rain, diffusiophoresis may

be significant. Greenfield (1957) has investigated radioactive fall-out using a model. It was found that there was a pronounced minimum of particles scavenged in the range 0.1 to 1.0 microns but below 0.01 microns and above 10 microns nearly all the particles were removed. For particles 2 microns radius rainfall rate 2.5mm per hour, 90% of the total activity was removed, the cloud drops in each case were 20 microns. Particles below 1 micron were assumed to mix with the water cloud before rain started. These could then be scavenged by cloud droplets by a coagulation process and hence removed by the rain.

2.6 The effect of electric forces in scavenging

Wang, Grover and Pruppacher (1978) have modelled the collision efficiencies of raindrops and aerosol particles for different radii, taking electric charges and different humidities into account. Drop sizes 8.6, 18.6, 30, 42, 72, 106, 173 microns were used. The electric charges were claimed to be realistic. The efficiencies decreased quite markedly from about one to one five-hundredth for particle radius 0.1 microns. The effect of electric charge was quite marked, the tendency being to increase collection efficiency up to two orders of magnitude over the range 0.005 to 1.0 microns, the resultant efficiencies being as high as one for 8.6 microns drops but about 0.005 for 173 micron drops. The effect of electric forces on scavenging has been investigated by Dayan and Gallitz (1975) experimentally and they found that the collection efficiency was increased by 1 to 2 orders of magnitude over uncharged drops. May (1958) investigated the washout of lycopodium spores by rain and found charges of about 6×10^{-4} esu. Smith (1955) showed that charging on drops varies from 10^{-1} to 5×10^{-5} esu. Adam and Semonin (1970) looked at submicron scavenging and found that for a charged drop radius 200 microns, collection efficiency was about 10%, though

these charges may be high. For larger drops and uncharged drops this was negligible. The experimental work was carried out using bacillus spores. Charged ice crystals are also thought to contribute to submicron scavenging.

3. THE INFLUENCE OF METEOROLOGICAL PARAMETERS ON THE SIZE DISTRIBUTION OF NATURAL AEROSOL PARTICLES

3.1 Introduction

Ground-based measurements of the concentration and size distribution of aerosol particles in the range 0.5 to 10 μ m diameter were made at both Manchester and Great Dun Fell, using a Royco optical particle counter. They are described and interpreted in terms of the prevailing meteorological conditions.

A considerable amount of data exists defining the aerosol particle spectrum for a wide range of localities and meteorological conditions, though the manner in which the particulate spectrum has been influenced by the synoptic meteorological situation has usually received little attention. However, some studies have investigated the relationship between prevailing meteorological conditions and bulk concentrations of pollution.

In an attempt to encompass both of these approaches we have undertaken preliminary studies of the influence of meteorological parameters on the size distribution of large/giant aerosol particles ($0.5\mu\text{m} < \text{particle diameter} < 10\mu\text{m}$) for prolonged sampling periods. Measurements have been made at two widely different locations, one urban with its concomitant anthropogenic sources of pollution, and one remote, rural, high level. The meteorological data used was routine measurements available from the national weather service.

3.2 Measurement Sites, Experimental Estimates and Data Sources

The city of Manchester is situated in a basin surrounded by the Pennine Hills to the north, east and south reaching to ~500m a.s.l in the north and east. To the west the land is comparatively flat encompassing the Lancashire plain and Mersey estuary. Measurements of particle size distribution were made from the roof of the main building of UMIST campus which is

adjacent to the city centre. By contrast the UMIST field station on Great Dun Fell (GDF) in Cumbria is 847m asl and located on an isolated peak on the Pennine ridge above the Eden Valley which runs northwest/southeast.

The particle size distribution in the size range 0.5-10 μ m was monitored using a Royco model 225 optical particle counter with model 507 digital display and autoscan facility and model 127 digital printer. The autoscan facility permits sequential monitoring of particle concentrations in the five size ranges 0.5-0.7, 0.7-1.4, 1.4-3, 3-5 and 5-10 μ m diameter. Prior to its initial use the counter was calibrated using polystyrene latex spheres.

Meteorological data was obtained from the Manchester Weather Centre (MWC) from their detailed logs together with upper air data taken from the Daily Aerological Record (DAR) for Aughton, about 50km west of Manchester and other sonde stations around Britain when appropriate. By using the 6-hourly synoptic charts in the Daily Weather Report (DWR) trajectories of different airmasses could be obtained as well as information about the location of frontal systems. Because there is no radiosonde station near GDF a combination of ascents at Shanwell, Aughton and Long Kesh together with a consideration of air trajectory, can be used to infer the structure of the atmosphere in the vicinity of the mountain.

Aerosol data for the periods 9-27 February 1978 in Manchester and 10-16 September 1977 at Great Dun Fell are discussed herein. The period 10-21 February 1978 was dominated by a polar continental airstream and 22-27 February by a mainly maritime regime. The GDF data is influenced by a basically unstable polar maritime flow with a strong subsidence inversion at about 900mb close to the level of the Great Dun Fell summit.

3.3 Results and Discussion

(1) Period 10-16 February 1978 (Manchester data)

Throughout this week a polar continental airstream prevailed although three subtle changes of airmass can be detected. From 00h 10/2 until 00h 12/2 the airstream had its source region in Northern Finland (airmass 1), thereafter the source was mainly Western Russia (airmass 2) with the airflow arriving at Manchester from a mainly N-NE direction. As a depression moved north from the Mediterranean into the North Sea late on the 14th, Manchester came under its cyclonic influence. Air circulating around this system came mainly from the near continent and arrived from a mainly east to south-easterly direction (airmass 3). The trajectories of these airmasses are illustrated in Figure 1.

From the diagrams of the aerosol counts against time (Figs.2 & 3) it is apparent that the long range trajectory is only of secondary importance in determining the levels of various particle sizes on these occasions. Shorter range trajectories appear to be much more significant. Broadly the counts in 0.35-0.7 μ m radius range show an increase throughout the week (10/2-16/2) especially after 13/2 and this is well correlated with the change in local wind direction in Manchester rather than changes in the source of the air. Early in the week the direction is mainly 010 $^{\circ}$, and has backed to 320 $^{\circ}$ on the 13th. Thereafter there is a steady veer through North and East to 110 $^{\circ}$. Figure 4 illustrates the changes in count of the 0.35-0.7 μ m range with wind direction. This particular size range of particle has its highest concentration when the air has come from the industrial West Midlands of England. However giant particles, eg, 2.5-5.0 μ m radius range do not show this effect and this would suggest a local source

for these particular aerosols, reflecting the shorter airborne lifetimes of the larger particles.

The stability of the boundary layer proves very important in determining aerosol concentrations by affecting the depth to which turbulent mixing either by convection or shear can occur. Values for the Richardson number have been computed from radiosonde data at Aughton for the relevant periods: the Richardson number (Ri) is given by

$$Ri = \frac{g}{\bar{\theta}} \frac{\partial \theta}{\partial z} / \left(\frac{du}{dz} \right)^2$$

where g = gravitational acceleration

$\bar{\theta}$ = mean potential temperature of the layer

$\frac{\partial \theta}{\partial z}$ = potential temperature gradient of the layer

$\frac{du}{dz}$ = shear in the particular layer.

This parameter effectively measures the relative production rates of turbulent kinetic energy by buoyancy (free convection) to shear (forced convection). $Ri > 0$ is characteristic of stably stratified layers while $Ri < 0$ implies instability and thus buoyant motions will be present.

Figures 5 and 6 show the relationship between the Richardson number and the aerosol counts for both 3.5-0.7 μ m and 2.5-5.0 μ m size ranges. The data was plotted for airmass types 2 and 3. The highest counts for small particles are found to coincide with periods of strong inversion conditions (large and positive Ri) as emphasized by data on 14/2 where a very intense nocturnal inversion is present. During this particular week the stability of the lowest layers decreased markedly during the day (eg on 14th as shown by the Aughton ascent data in Figure 7) as solar heating initiated convection and subsequently concentrations of small particles decreased sharply. The 14th is a prime example

of this effect when following dispersal of the inversion convection was established up to 750mb and snow showers were reported. For the large particle sizes ($2.5\mu\text{m}$ radius) there is a negative correlation between concentration and Richardson number. The implication is that during periods of large and positive Richardson numbers (high stability and thus little turbulence) the gravitational settling of these particles is important and sufficient to account for the low concentrations of these particles in the early mornings of these days. Further reference to the diurnal variation of the particle sizes will be made later on in this report.

(11) Period 21-27 February 1978 (Manchester Data)

This period is rather complex meteorologically with a fundamental change of synoptic type early in the period. From 00h 21/2 to 06h 22/2 there is a gradual encroachment of Atlantic air behind a series of weak occlusions and true maritime air does not arrive until the 24th. Some of the airstream trajectories are illustrated in Figure 8. After the 24th a series of troughs and fronts in the generally S-SW flow brought periods of rain or showers. Generally however the concentrations of particles in all size ranges are little changed from those of a week earlier in the continental regime, again emphasising the importance of short/medium range trajectory.

As with the previous week's data, the concentration of aerosols is closely associated with stability for a given air-mass. An intense low-level frontal inversion provides an extremely effective barrier for the dispersion of particulate matter around 00h on 22/2 while a nocturnal inversion on the 27th is also responsible for high concentrations of small particles. As for the 14th, the 27th shows similar characteristics with deep convection occurring once the inversion has dispersed

eventually producing rain showers. The extremely low concentrations of the 24th are similarly associated with a very deep convective mixing layer, and thus large turbulence giving rise to comparatively large concentrations of giant particles.

From 00h on 23/2 the smaller particle concentration rises and is closely associated with a veering of the wind from E to S, with the boundary layer stability remaining essentially unchanged, thus confirming the significance of medium range air trajectories; the industrial Midlands is the likely source again.

Figure 9 illustrates the diurnal variation of the particle concentrations during the period of data at Manchester. For particle sizes from $0.25\mu\text{m}$ up to $1.5\mu\text{m}$ radius there is little evidence of any diurnal variation but there are strong variations for particles upwards of $2.0\mu\text{m}$ radius. The reason for this is the diurnal variation of turbulence in the boundary layer, and therefore Richardson number. As convection becomes established in the early morning after about 0700hrs, the turbulence in the boundary layer increases (Ri decreasing) eventually reaching a maximum intensity at about 1500hrs. Thereafter convective turbulence declines and the ability of the turbulent fluctuations to retain giant particles aloft is diminished. Because these giant particles have sufficiently large terminal velocities ($\sim 1.0\text{cm s}^{-1}$), gravitational settling becomes an important process in determining their concentration and a minimum concentration is observed around 0600hrs which is the time of minimum turbulence, ie, maximum Richardson number. In summary then, the diurnal variation of stability in the lowest layers will be the most fundamental factor in determining the concentration of giant particles.

To test this conclusion, correlation coefficients of particle size concentrations against various meteorological

parameters were calculated and the results are presented in Table 1. Illustrative graphs of count against meteorological parameter are presented in Figures 10 and 11. Table 2 examines the significance of these calculations. Strong correlations between wind speed, temperature and Ri for the giant particles provide more evidence of the effect of the diurnal variation of turbulence. For the small particles there is a strong positive correlation with the Richardson number, but the large positive values of Ri are generally associated with intense low level inversions either frontal or as is more usual, to radiation cooling. Lower values of Ri are associated with greater mixing depths in convective situations. Elsom (1979) provides evidence of low level trapping by a temperature inversion which because of its longevity gives rise to anomalously high levels of pollution in Manchester.

(iii) Period 16-20 September 1977 (GDF data)

A weak trailing cold front, associated with a deep depression over Western Russia, lying from Essex to Northern Ireland at 00h 16/9 moved away slowly South-West and had cleared the British Isles by 00h 17/9. Meanwhile a large anticyclone situated over Iceland at 12h 15/9 had moved slowly South-east to lie between the Faeroe Islands and Shetlands by 12h 17/9 (Figure 12). Thereafter the synoptic situation remained essentially unchanged with the anticyclone becoming stationary and keeping much the same intensity. The effect of these synoptic changes was that the winds remained north-easterly throughout the period although subtle changes of trajectory occurred. The airstream on the 16/9 and 17/9 had its origin over Greenland and had reached GDF around the eastern and south-eastern flanks of the anticyclone. By the end of the period as the anticyclone had slipped south-

eastwards, the airstream was coming from an Atlantic origin. As these air masses came south across the North Sea, heating by the oceans had caused them to become unstable in their lowest layers which were capped by a strong subsidence inversion associated with the anticyclone. The trajectories are shown in Figure 13.

Observations of a full stratocumulus cloud cover at the east coastal stations of Scotland and Northern England are characteristic of this effect, thus particulate matter will be well mixed in the layer up to the inversion.

A graph of particle concentration against time (Figure 14) illustrates large variations which were of similar magnitude and were in phase for all the size categories. As stated previously the meteorological conditions are essentially unchanged and thus changes of airmass cannot account for the large fluctuations, eg, 00h 19/9 to 14h 19/9. However, changes in the height of the subsidence inversion are very noticeable from radiosonde ascents at Stanwell and Aughton. From a consideration of air trajectory and distance from the centre of the anticyclone it is thought that these two stations provide a representative sounding of the air in the vicinity of Great Dun Fell. Since the meteorological data ascents are only made at 00h and 12h during the day so the concept of mixing height has to be employed to extrapolate values of the inversion height at times between the standard observations. The idea of mixing height was proposed by Holzworth (1967) and the maximum mixing height is derived as the intersection of the dry adiabat extended from the maximum temperature to the sounding at 12h. The minimum mixing height is the intersection of the dry adiabat from the minimum temperature (plus 5.0°C in urban areas to account for heat island effects) to the sounding at 00h.

Figure 15 illustrates the variation of the inversion height with time. The inference to be drawn is that the periods of very low particle count are when the inversion falls below the mountain top and the site is in a region where the air has come from high up in the troposphere by subsidence. Higher particle concentrations are always observed during the day and it is likely that turbulent mixing caused by surface convection from the valley pushes the inversion above the mountain top with the inversion providing an effective bar against further dispersal. Consequently when convection dies away again in the evening the inversion falls below the mountain top and the site once again enters the subsidence zone. This effect occurs three times during the period. (The sampling period was terminated when the local standby diesel generator was activated, resulting in the sudden increase in particle concentrations on 20/2) Ascents at both Shanwell and Aughton indicate a well-mixed layer below the inversion during the day with a super-adiabatic lapse rate established near the ground and a dry adiabatic lapse above it, up to the inversion.

Throughout the period the winds at 900m at both Shanwell and Aughton remain less than 20kts from the North-east. If the wind is strong it is observed that inversion layers can be lifted bodily over GDF because the flow has sufficient energy to do the work required. For lighter airflows this is not necessarily the case and the inversion can fall below mountain top level particularly when a strong subsidence inversion is present. Thus it may be deduced that the variation in the height of the inversion layer is sufficient to account for the fluctuations observed in the aerosol spectra.

TABLE 1
Correlation Coefficients for Manchester Data

Particle Size (μm radius)	Parameter	Correlation Coefficient (r)
0.25-0.35	Wind speed	+ 0.42
	Relative Humidity	- 0.27
	Temperature	+ 0.33
2.5-5.8	Wind speed	+ 0.87
	Relative Humidity	- 0.88
	Temperature	+ 0.95
0.35-0.7	Richardson number	+ 0.94
2.5-5.0	Richardson number	-0.85
0.25-0.35	Rainfall amount	-0.17
1.5-2.5	Rainfall amount	-0.43
2.5-5.0	Rainfall amount	-0.47

TABLE 2

Parameters	Correlation Coefficient (r_A)	Probability of r being by chance > r_A
Wind speed, relative humidity and Temperature	0.62	0.10
	0.84	0.01
Richardson number	0.81	0.05
	0.88	0.01
Rainfall amount	0.45	0.10
	0.53	0.05

REFERENCES

- Adam J R and Semonin R G 1970 "Collection efficiencies of raindrops for sub-micron particles" Precipitation scavenging symposium June 1970 Ref. CONF-700601 p151-160
- Beard K V 1976 J.Atmos.Sci., p851 (May)
- Beard K V and Grover 1974 J.Atmos.Sci., 31, 543-550
- Dayan M and Gallitz T 1975 J.Atmos.Sci., 1419-1429
- Elsom D M 1979 Weather, 34, 277-286
- Goldsmith P,
DeLafield H J and
Cox L C 1963 Quart.J.Roy.Met.Soc., 89, 43-61
- Greenfield S M 1957 J.Met., 14, 115
- Junge C E 1963 Geophysical Series, Vol 4m Academic Press, New York "Air Chemistry and Radioactivity"
- Holzworth G C 1967 J.Appl.Met., 6, 1039-1044
- Ketz U and Kocmond C 1973 J.Atmos.Sci., 30, 160-165
- Kerker M and Hampl V 1974 J.Atmos.Sic., 31, 1318-1322
- Langmuir I and
Blodgett K 1945 "Mathematical investigation of raindrop trajectories". Gen.Elec. Res.Lab., Rep. Schenectady, RL/225
- May F G 1958 Quart.J.Roy.Met.Soc., 84, 451
- Perkins R W, Thomas C W
Young J A and Scott B C 1970 Precipitation Conf., 1970, 69-97 "Incloud scavenging analysis from cosmogenic radio nucleide"
- Pitter R L 1977 J.Atmos.Sci., 34, 1796-1800
- Rantz W S and Wong J E 1952 Ind.Ing.Chem., 44, 1371-1381
- Rogers R R 1976 "A Short Course in Cloud Physics" Vol 84, Pergamon Press
- Slinn W G N and Hales J M 1970 Precip. Scavenging Conf. 411-422 "Phoretic processes in scavenging"
- Smith L G 1955 Quart.J.Roy.Met.Soc., 81, 23
- Sood H K and Jackson M R 1970 "Scavenging by rain and ice crystal Precip. Scavenging Conf. 131-136
- Storebo P B and Dingle A N 1974 J.Atmos.Sci., 31, 525-542
- Takahashi T 1976 J.Atmos.Sci., 269-275
- Turner J M 1955 Quart.J.Roy.Met.Soc., 84, 418-492
- Twomey S 1977 "Atmospheric Aerosols"

Wang P K, Grover N and
Pruppacher H R

1978 J.Atmos.Sci., 1735-1843

Weickmann H K and
Kampe A K

1953 J.Met., 10, 418-429

LEGENDS TO FIGURES

- Figure 1 Airstream trajectories to Manchester in the period 10th-16th February 1978.
1 10.2.78; 2 13.2.78; 3 16.2.78
- Figure 2 Particle concentration $N \text{ cm}^{-3}$ versus time in Manchester from 10th-16th February 1978 for channels 1-5.
radius range covered by channel
channel 1 0.25 - 0.35 μm
channel 2 0.35 - 0.7 μm
channel 3 0.7 - 1.5 μm
channel 4 1.5 - 2.5 μm
channel 5 2.5 - 5.0 μm
- Figure 3 Particle concentration $N \text{ cm}^{-3}$ versus time in Manchester from 21st-27th February 1978. Size ranges covered are those for Figure 2.
- Figure 4 Scatter diagram of wind direction θ versus percentage deviation from the average concentration D for channel 2. The average concentration is taken for the period 10th-16th February 1978.
- Figure 5 Richardson number Ri versus particle concentration $N \text{ cm}^{-3}$ for channel 2 for the period 06h 12.2.78 to 18h 14.2.78.
- Figure 6 Richardson number Ri versus particle concentration $N \text{ cm}^{-3}$ for channel 5 for the same period as Figure 5.
- Figure 7 Low-level radiosonde ascents data at Aughton on 14th February 1978. Dry bulb temperatures only at 00h (—) and 12h (---)
- Figure 8 Airstream trajectories to Manchester in the period 21st-27th February 1978.
1 22.2.78; 2 23.2.78; 3 24.2.78
- Figure 9 Diurnal variation of particle concentrations $N \text{ cm}^{-3}$ channel size categories 1-5 respectively
- Figure 10 Wind speed U , relative humidity ϕ and temperature T versus particle concentration $N \text{ cm}^{-3}$ for channel 1.

Figure 11 Wind speed U , relative humidity ϕ and temperature T versus particle concentration $N \text{ cm}^{-3}$ for channel 5.

Figure 12 Synoptic situation at 12h 17th September 1977.

Figure 13 Airmass trajectories to Great Dun Fell for the period 16th-19th September 1977

1 16.9.77; 2 18.9.77; 3 19.9.77

Figure 14 Particle concentration $N \text{ cm}^{-3}$ versus time for Great Dun fell from 16-19th September 1977. Channel sizes indicated are those for Figure 2.

Figure 15 Variation of the inversion height H with time from 16th-19th September 1977.

Height A is the level of Great Dun Fell summit.

Figure 16 Low-level radiosonde ascent data for Shenwell on 19th September 1977. Dry bulb temperatures at 00h (—) and 12h (---). The summit level of Great Dun Fell is indicated (A).

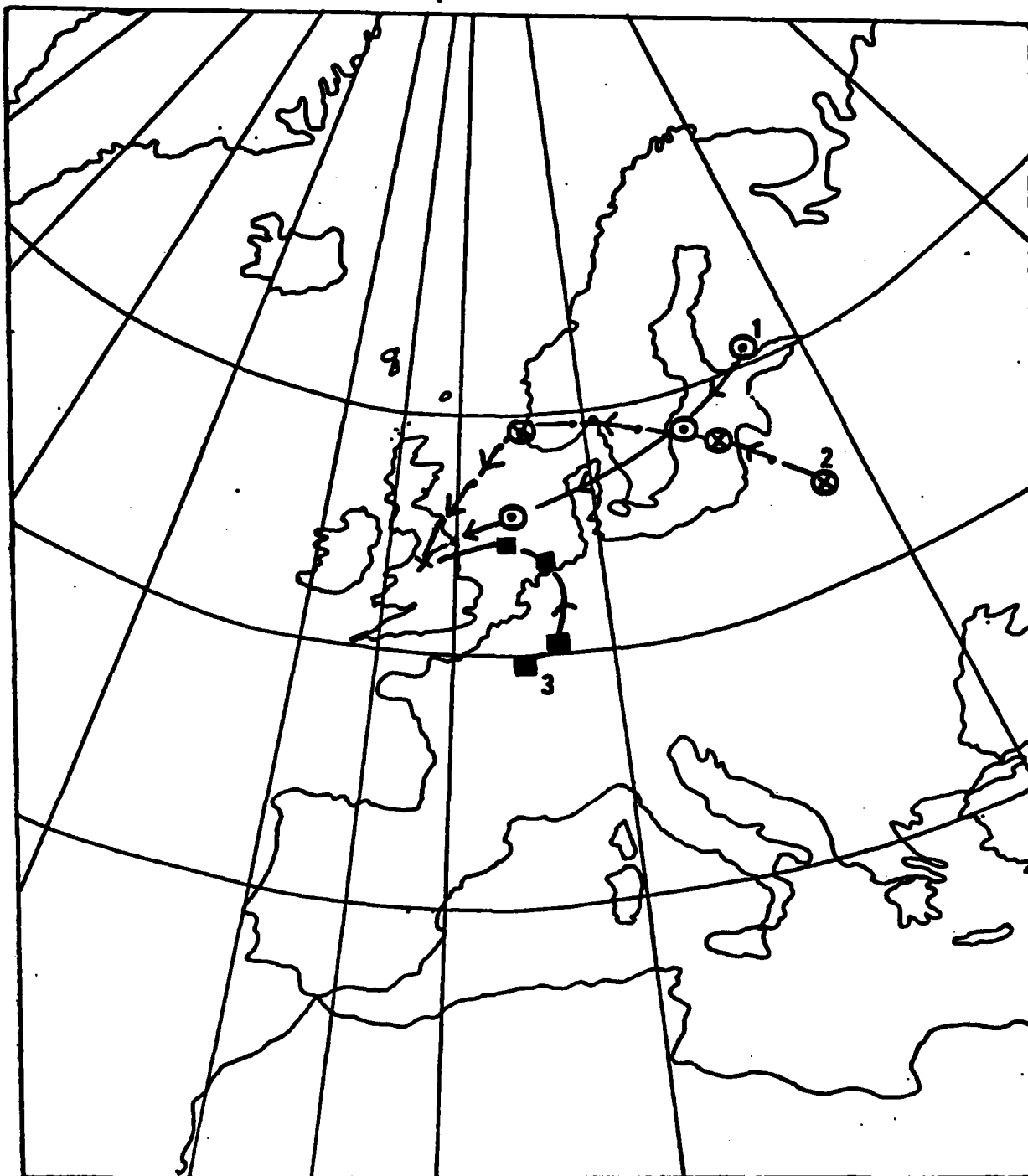


Fig. 1.

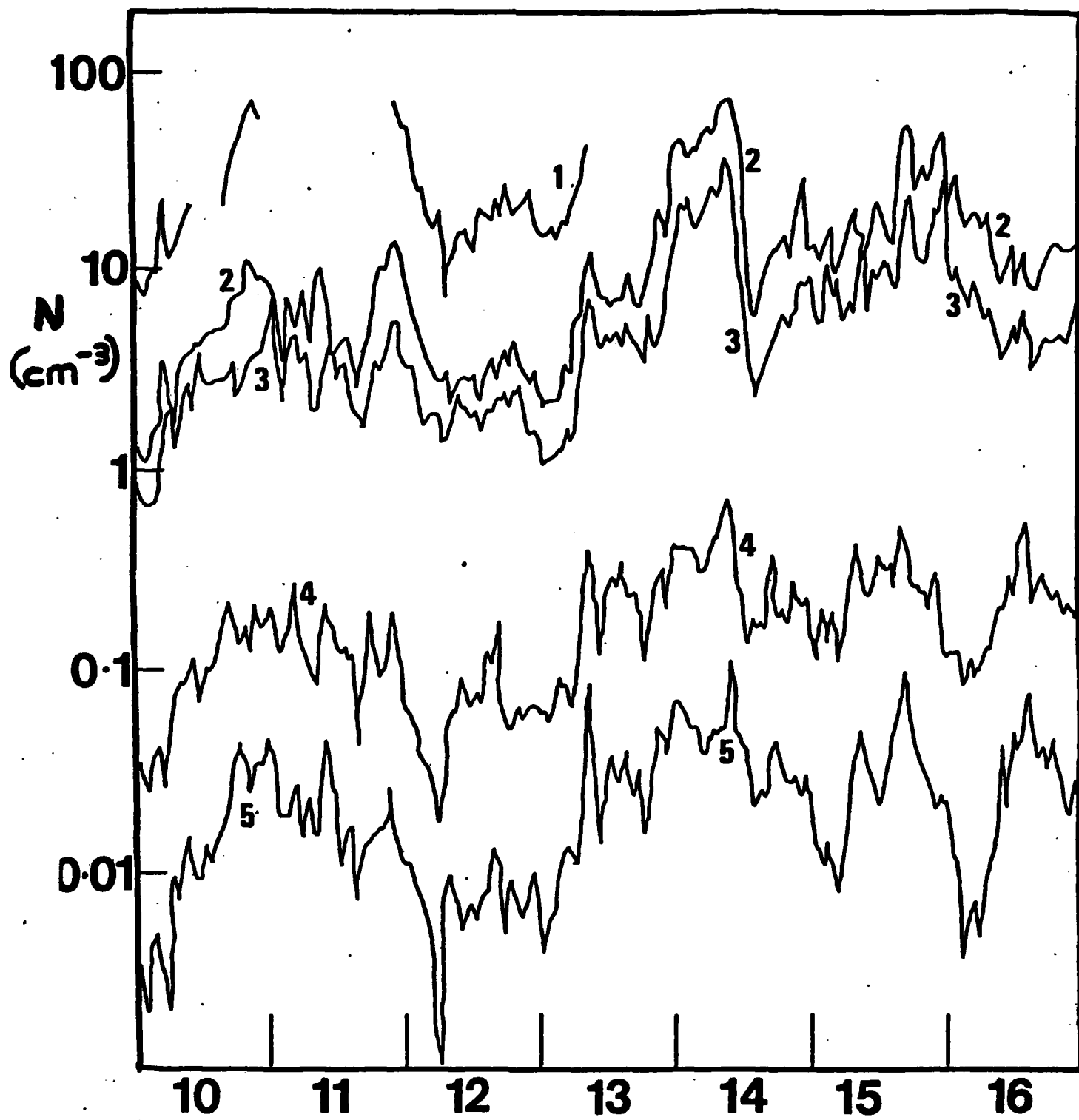


Fig. 2.

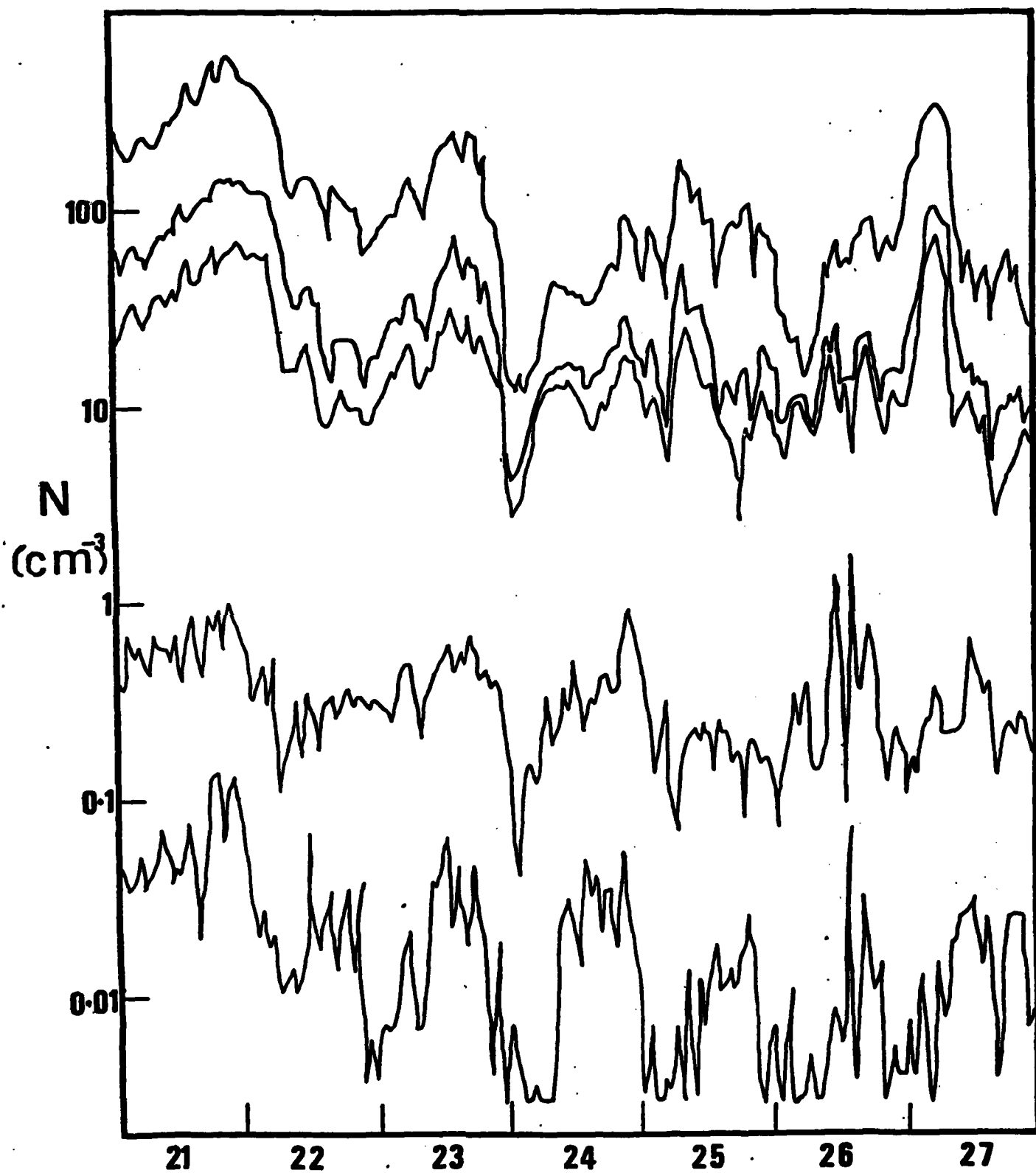


fig. 3

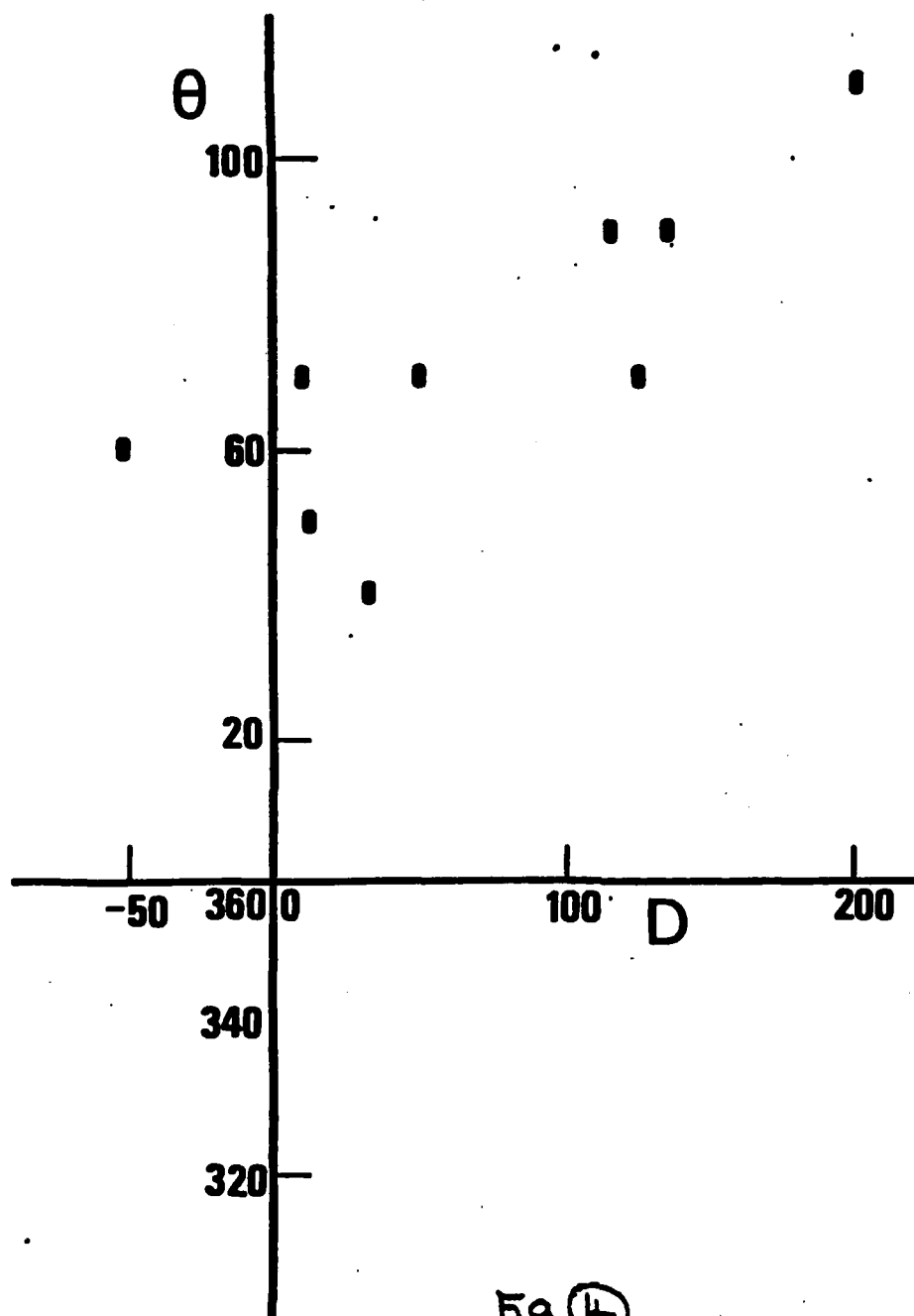


Fig ④

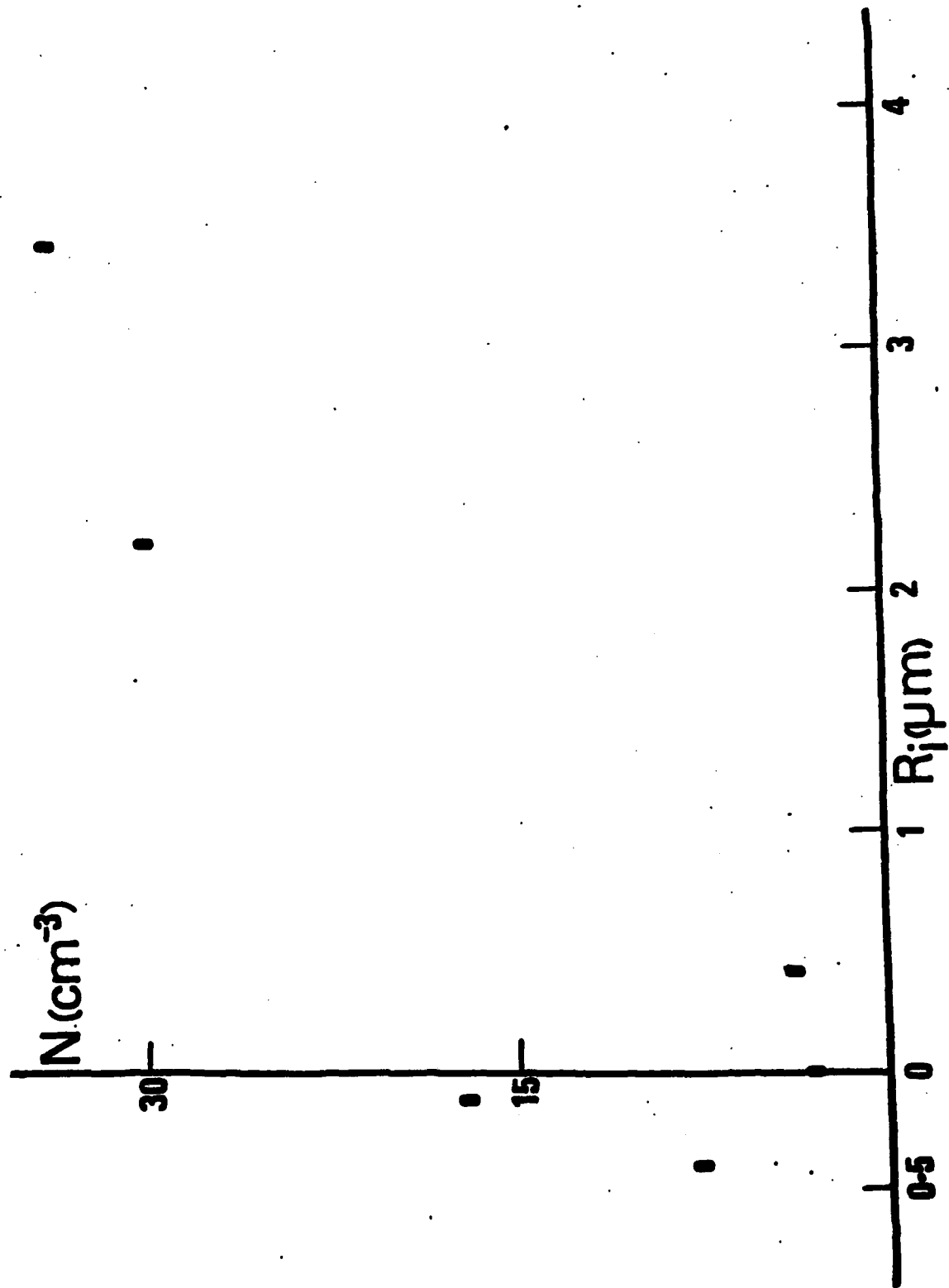


Fig. 5

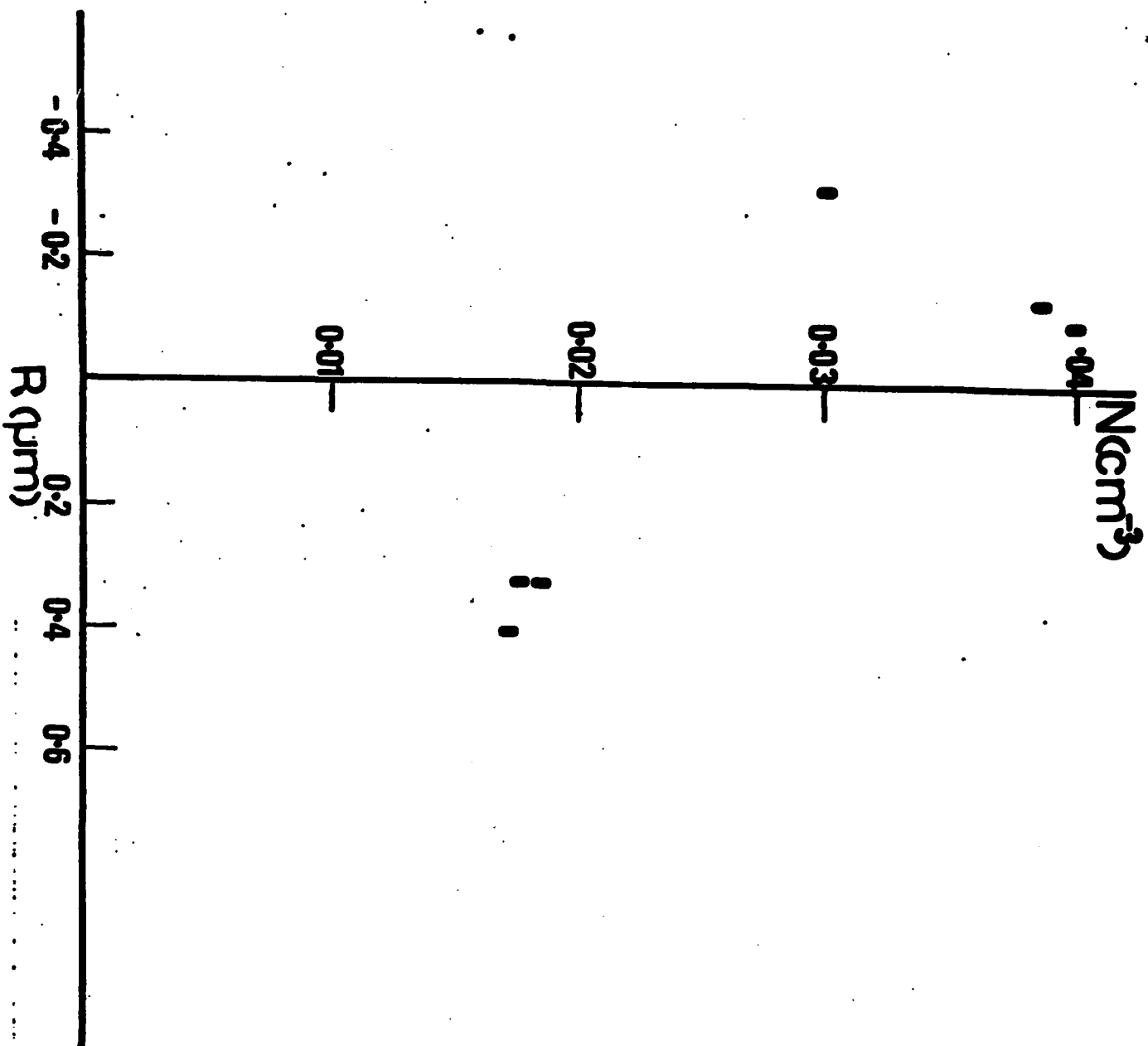


Fig. 4

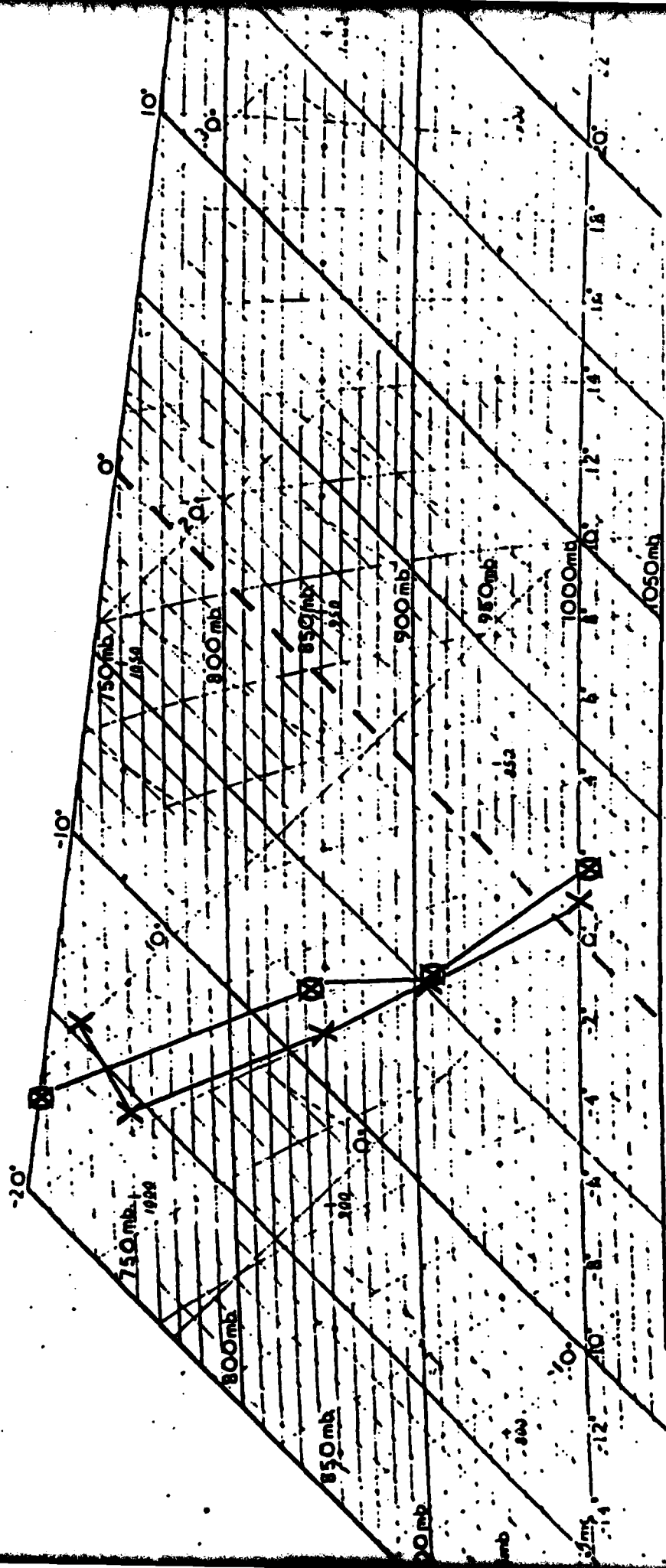


Fig. 7.

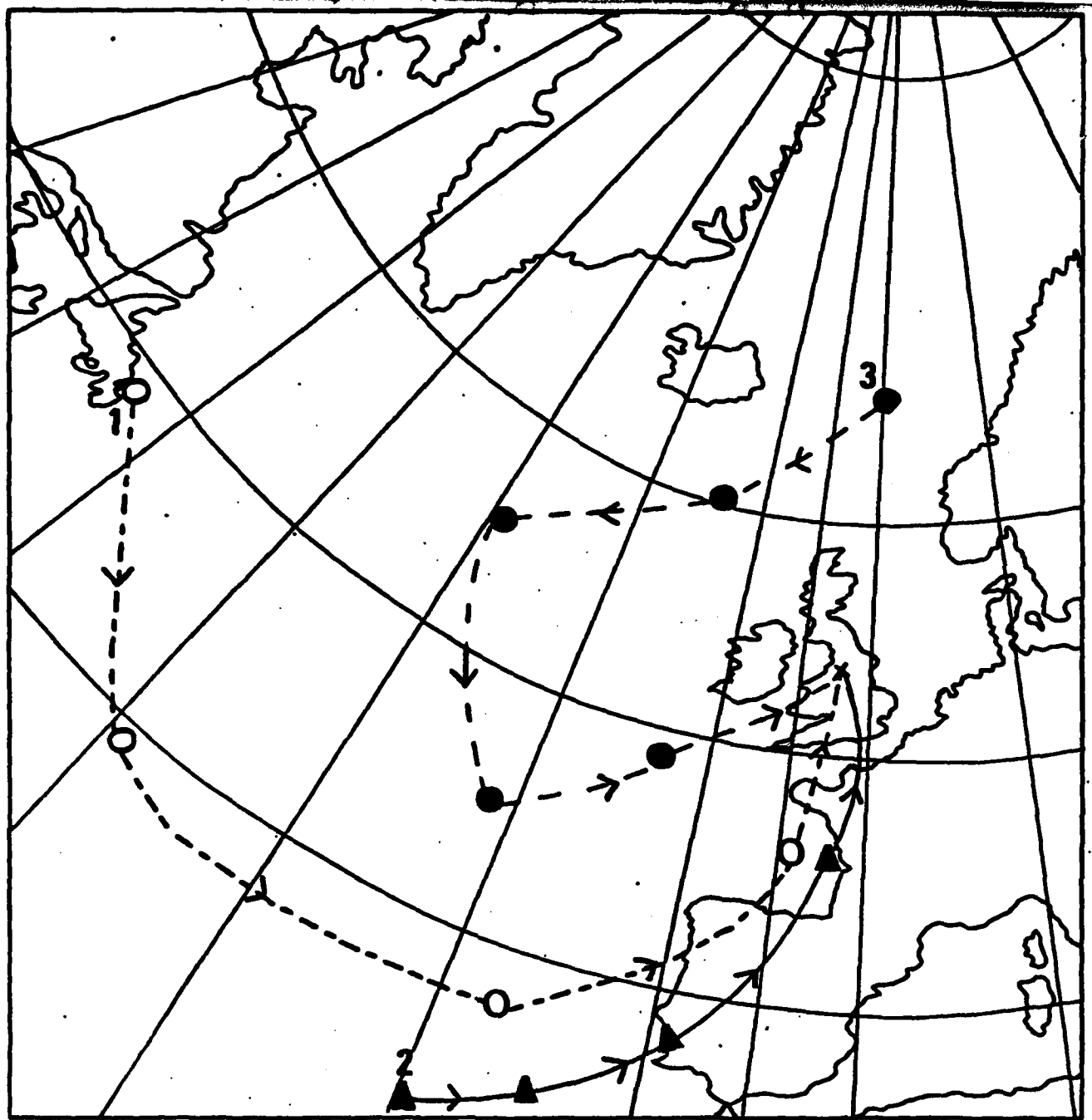


Fig. 8.

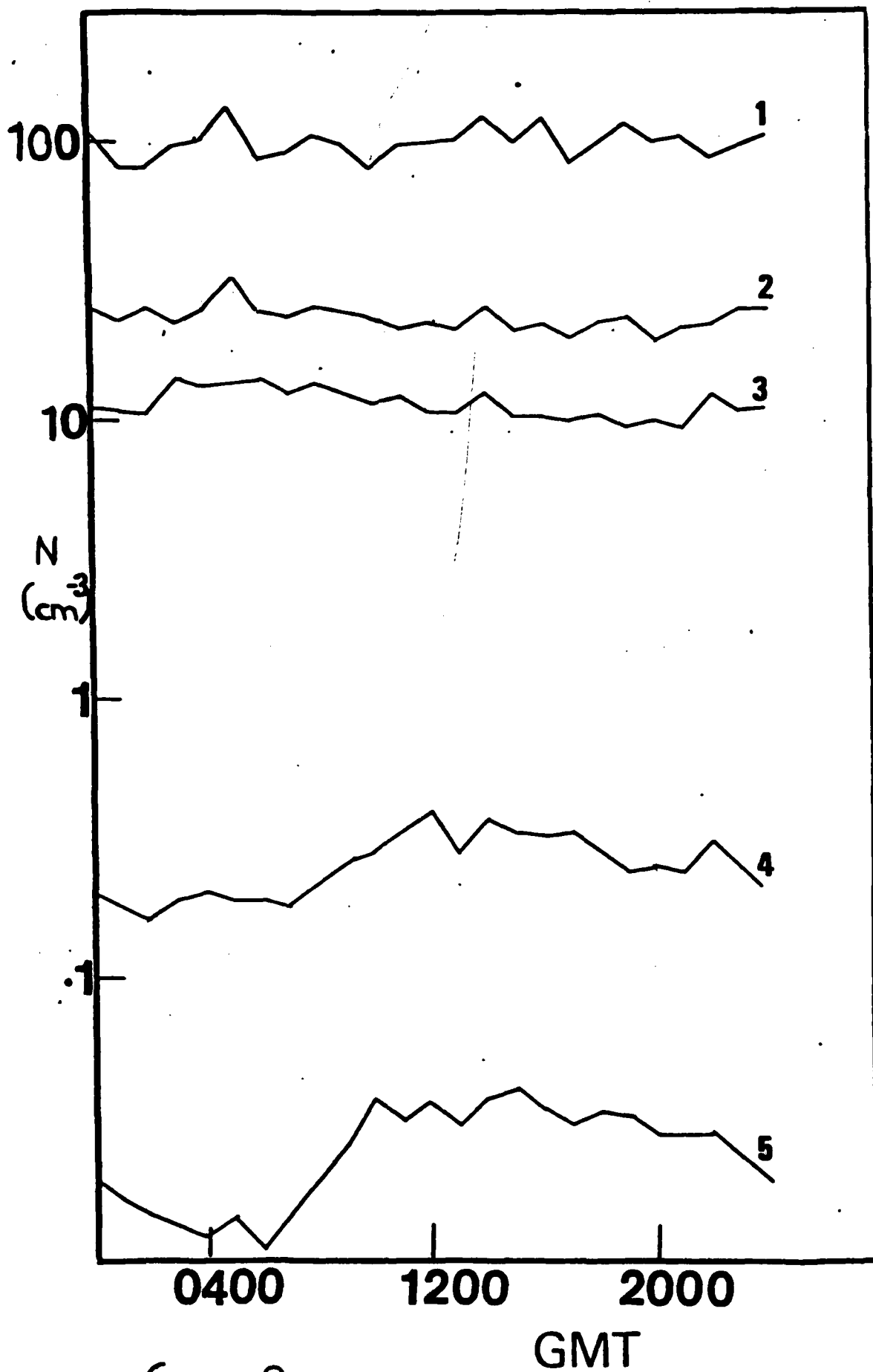


Figure 9

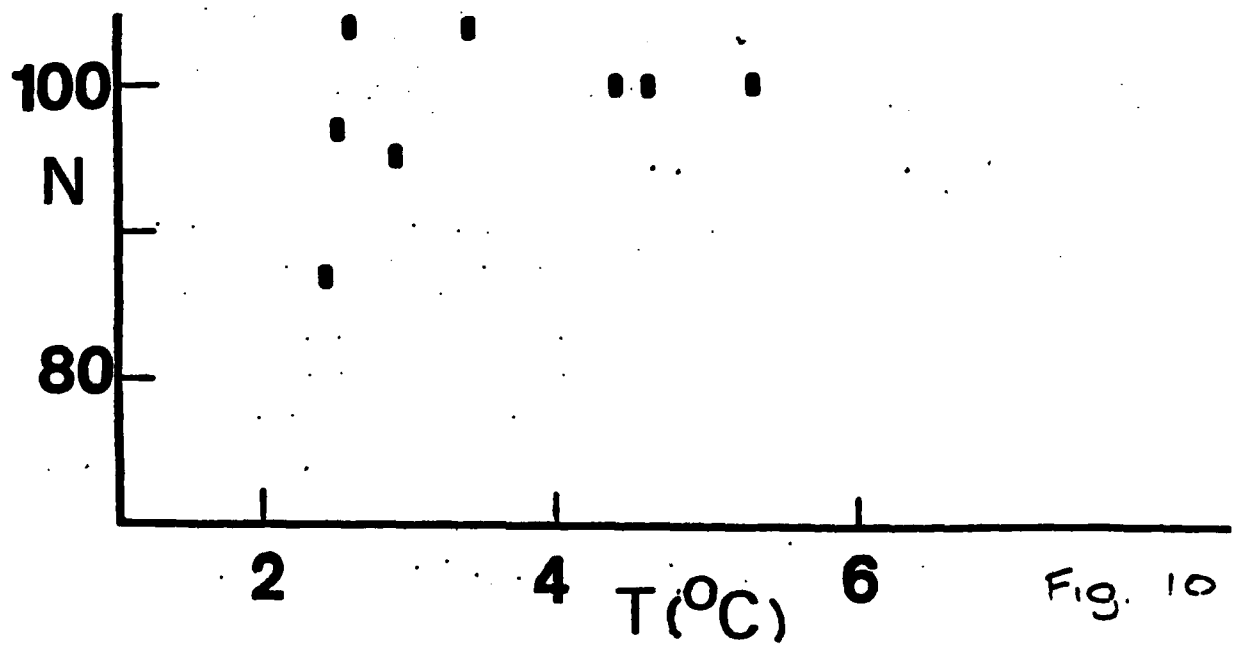
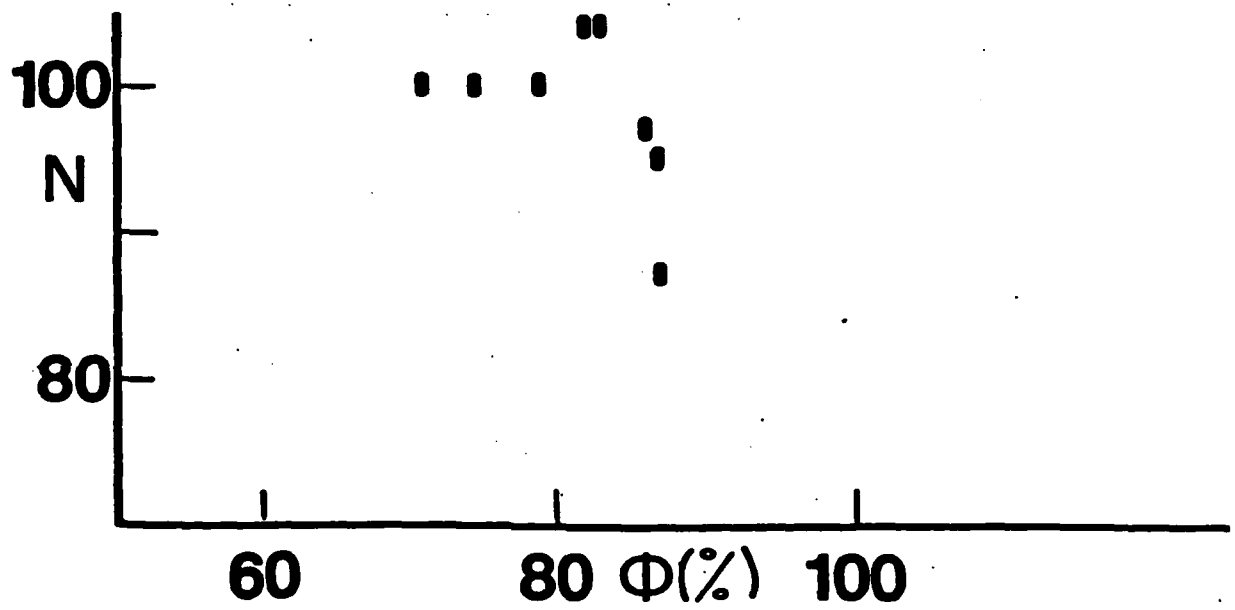
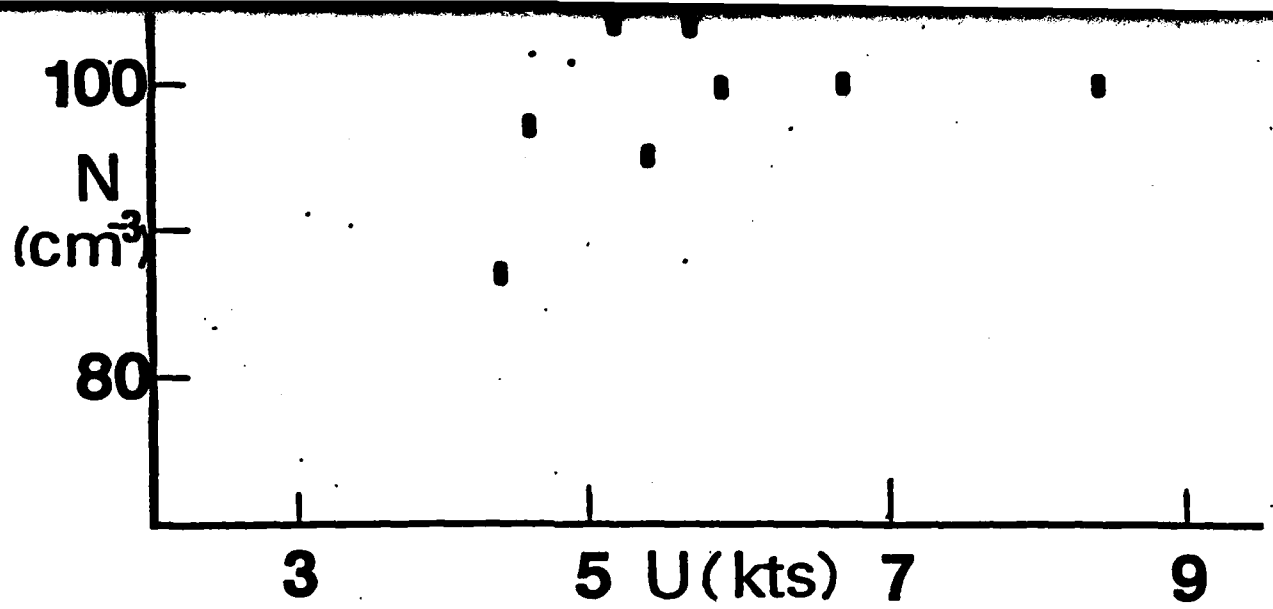


Fig. 10

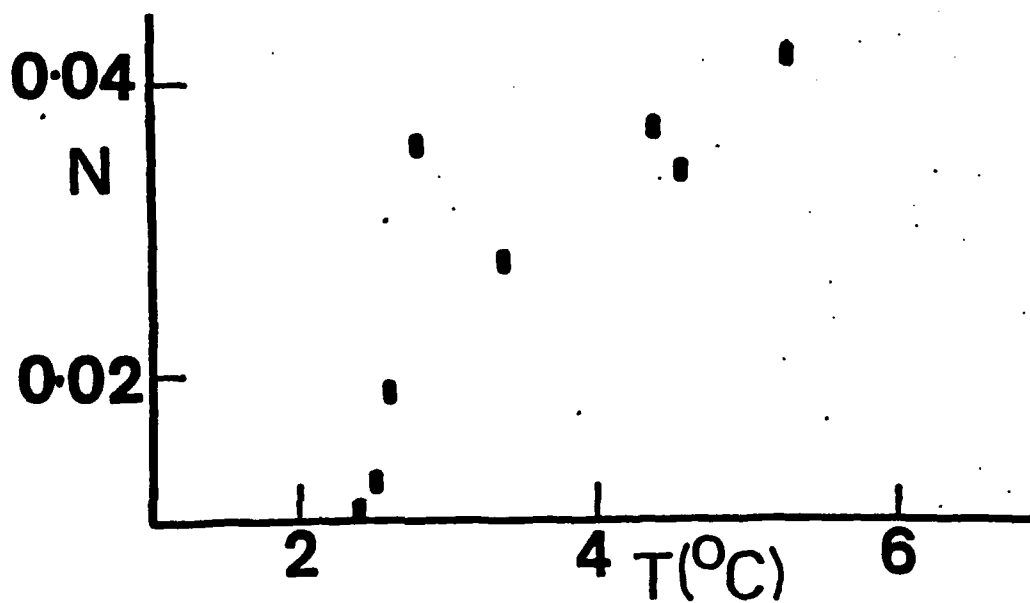
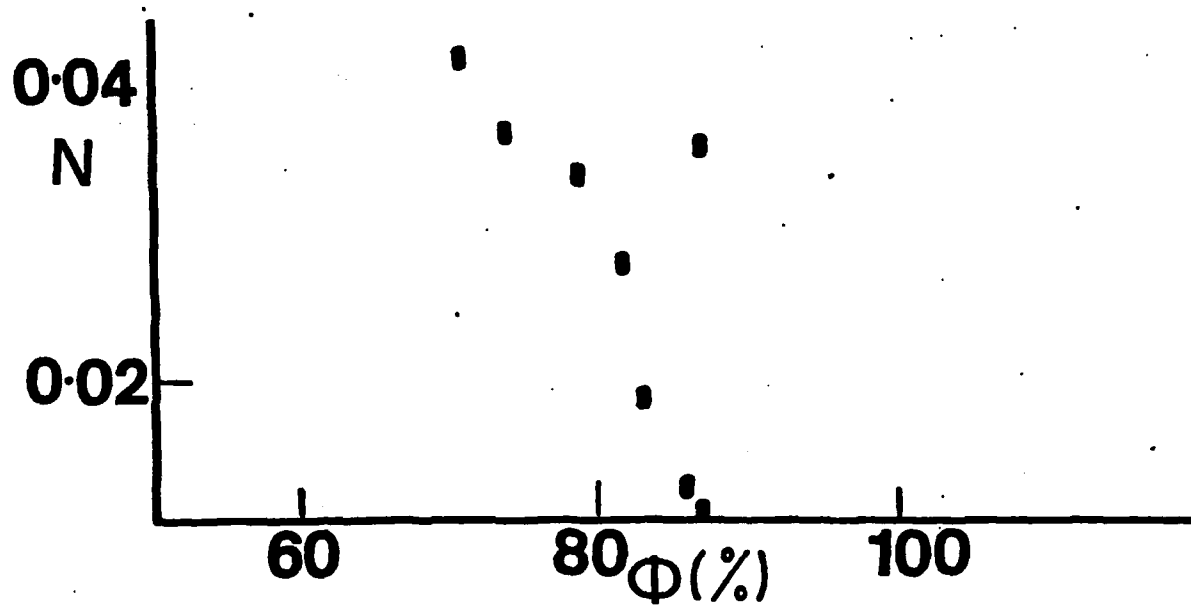
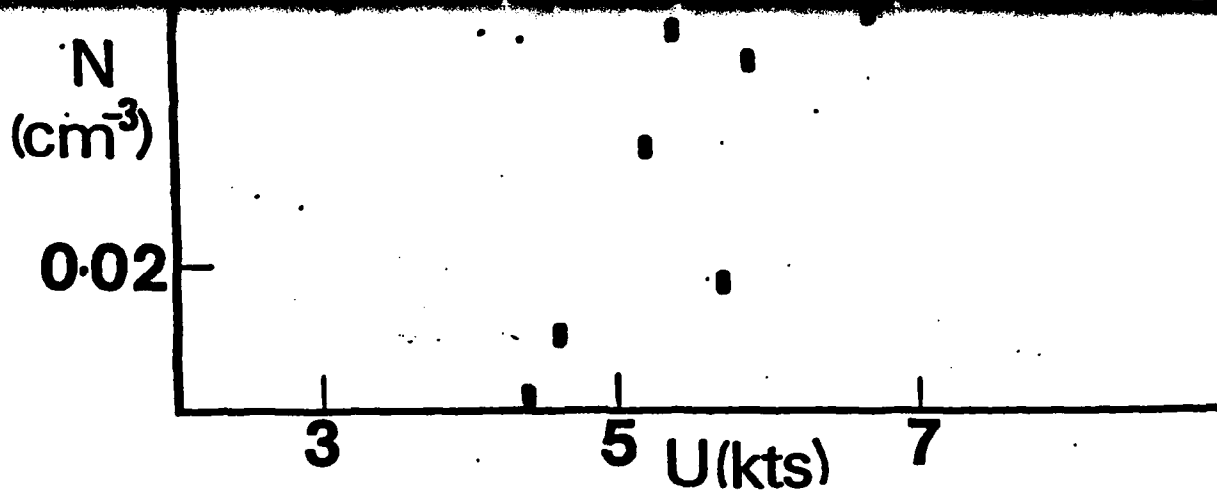
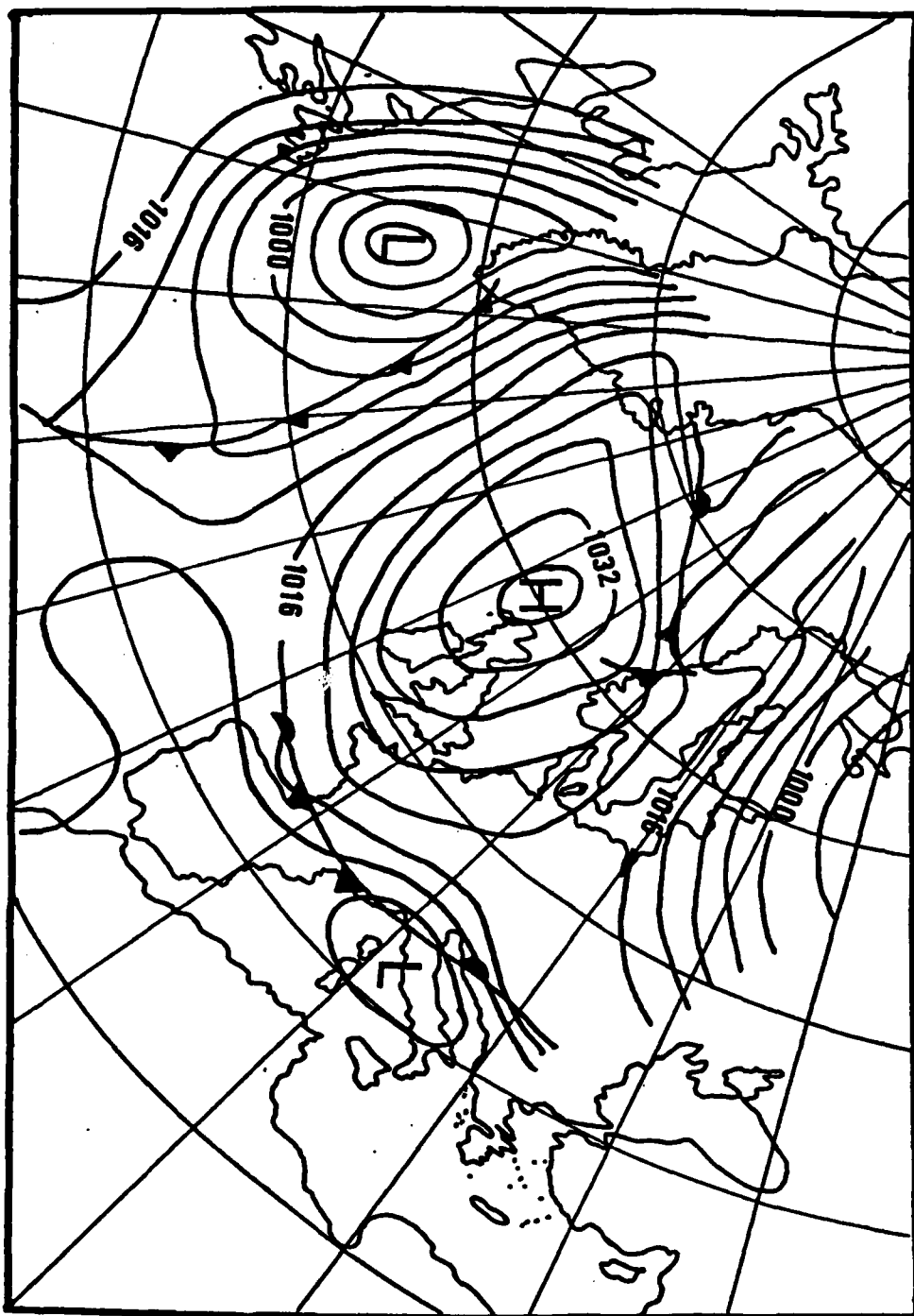


Fig. 11.

Fig. 12.



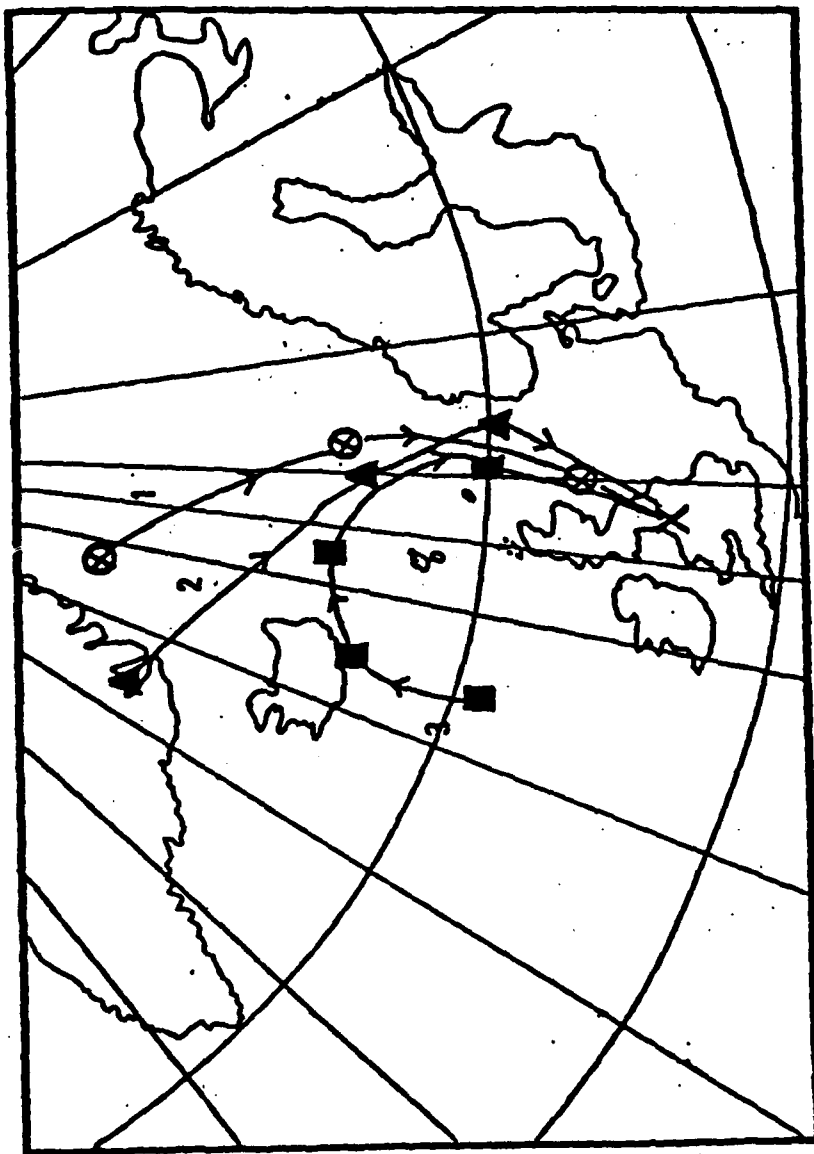
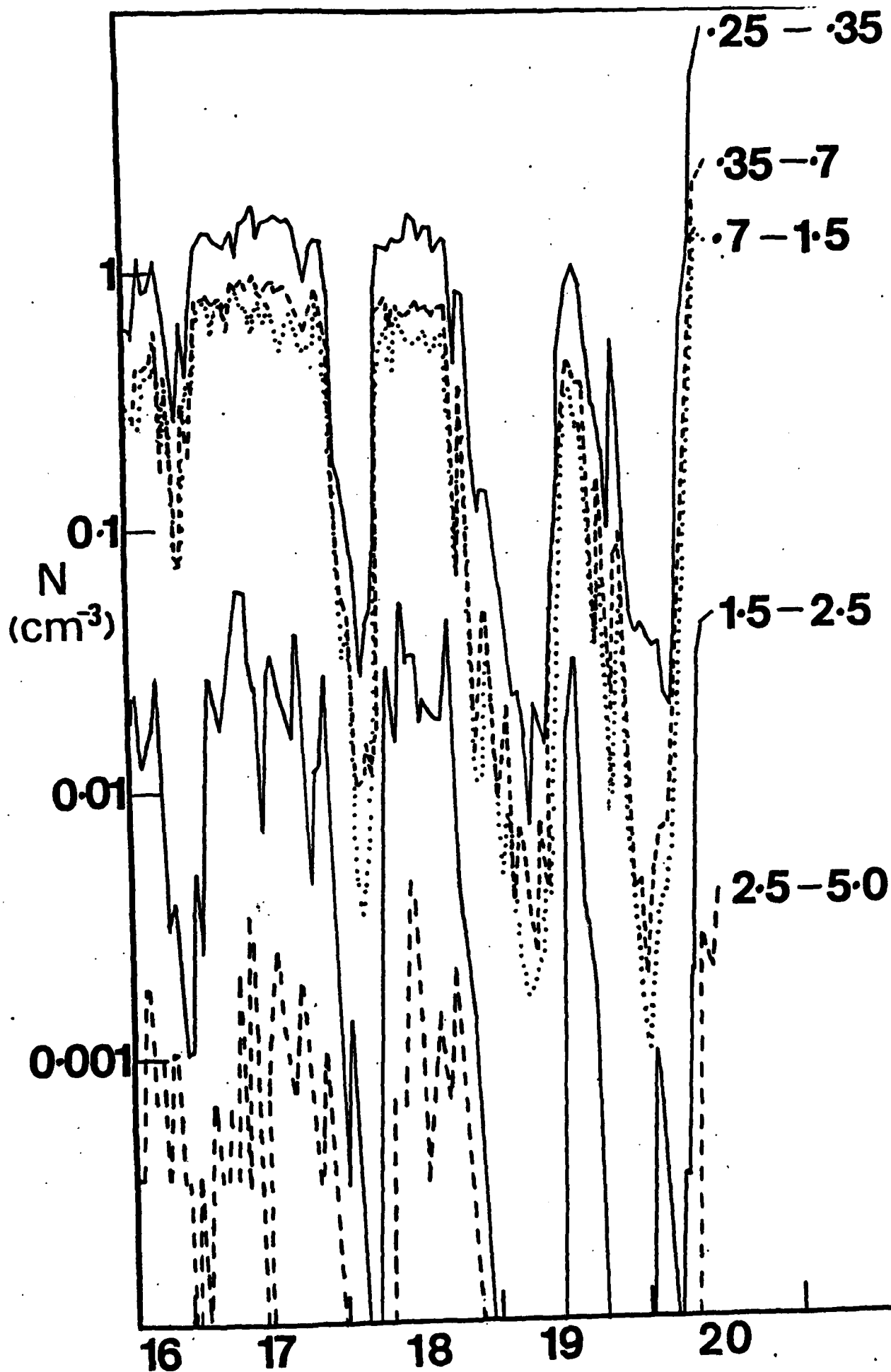


Fig. 13.

Fig(14)



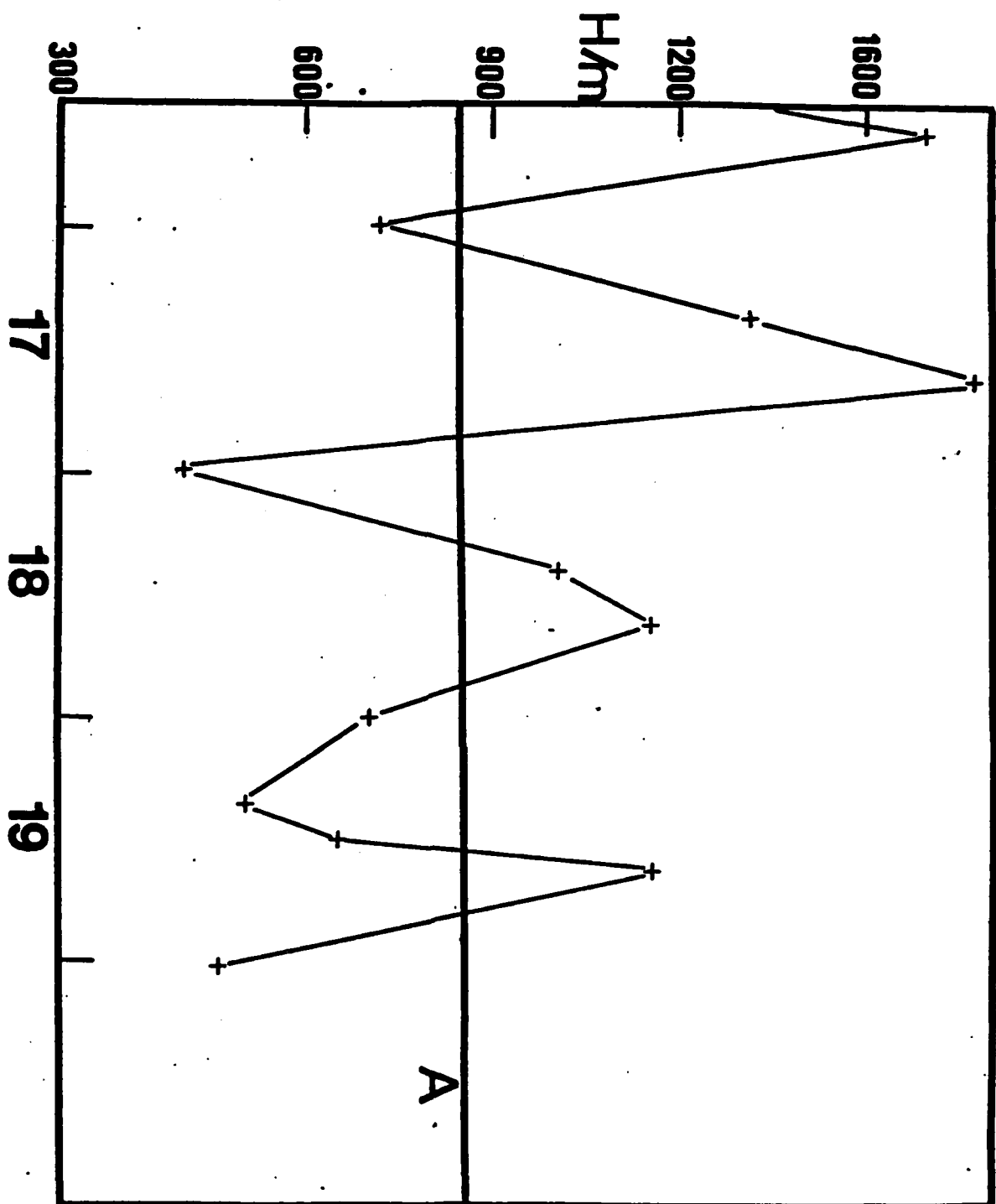


Fig. 15